A new process for the determination of the Soret coefficient of a binary mixture under microgravity

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ABSTRACT

In the presence of a gravity field or under microgravity, pure thermo-diffusion leads to very weak species separation in binary mixtures. To increase the species separation in the presence of gravity, many authors use thermo-gravitational diffusion in vertical columns (TGC). For a given binary mixture, the species separation between the top and the bottom of these columns depends on the temperature difference, ΔT, imposed between the two vertical walls facing each other, and the thickness, H, between these two walls (annular or parallelepipedic column). These studies show that, for a fixed temperature difference, the species separation is optimal for a thickness, H_{opt}, much smaller than one millimetre. The species separation decreases sharply when the thickness H decreases with respect to this optimum value. It decreases progressively as H increases with respect to H_{opt}. In addition, for mixtures with a negative thermo-diffusion coefficient, the heaviest component migrates towards the upper part of the column and the lightest one towards the lower part. The loss of stability of the configuration thus obtained leads to a brutal homogeneity of the binary solution.

The objective of this study in microgravity was to increase the optimum of species separation. For this purpose, the binary fluid motion was provided by uniform velocities imposed on the two walls of the cavity facing each other. This forced flow led to species separation between the two motionless walls of the cavity. In this case, the fluid motion generated in the cavity was not dependent on the imposed temperature difference, ΔT contrarily to the case of thermogravitational column. Under these conditions and for a given column of thickness H, there are three independent control parameters: ΔT and the two velocities of the walls facing each other. Using the parallel flow approximation for a cell of large aspect ratio, the velocity, temperature and mass fraction fields within the cavity were determined analytically. Thus the parameters leading to optimal species separation were calculated. The analytical results were corroborated by direct numerical simulations. The present paper thus proposes a new process for the determination of the Soret coefficient, the thermodiffusion coefficient and mass-diffusion coefficient.

1. Introduction

In natural environments and in industrial processes, most solutions are composed of several constituents. Most often, one of the constituents is of particular importance for industry, for the environment and/or for pharmacy. It is therefore important to be able to extract this component from the mixture in a natural, inexpensive way. Thermo-gravitational separation is mainly used to separate components with high added value (rare earths). Research involving binary and ternary mixtures and the measurement of their thermodiffusion coefficients is of great interest for industrial applications [1,2]. The Soret effect in liquid mixtures has recently been reviewed by Kohler et al. [3]. This important contribution includes numerous references and deals with thermo-diffusion in binary and ternary liquid mixtures. The most important experimental techniques currently used are also described and interpreted in this review. J.C. Legros [4] was at the origin of work carried out in microgravity in order to measure the Soret coefficients under good conditions and to compare the results with the experimental values obtained on the earth. His project planned to measure the Soret coefficients of twenty different systems. The hardware was described and the systems to be studied were listed. Chacha et al. [5] investigated the role of thermal diffusion phenomena on compositional variation in a binary mixture of methane and normal butane in the presence of a fluctuating acceleration field, in experiments on board the International Space Station (ISS). Shevtsova [6] proposed a new approach to simultaneously measure diffusion and the Soret coefficient. A binary mixture of water and isopropanol was chosen as the working liquid. The principle of the space experiments and

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Nomenclature

C	Mass fraction
D	Mass diffusion coefficient, $[m^2s^{-1}]$
D_T	Thermodiffusion coefficient $[m^2s^{-1}K^{-1}]$
f	Velocity Ratio of moving walls
g	Gravitational acceleration $[ms^{-2}]$
H	Height of the cavity $[m]$
L	Length of the cavity $[m]$
m	Mass fraction gradient $[m^{-1}]$
Le	Lewis number
P	Pressure $[Pa]$
Pe	Péclet number
Pe_m	Mass Péclet number
Pr	Prandtl number
Ra	Rayleigh number
S_T	Soret coefficient
t	time
T	temperature
T_h	hot temperature
T_c	cold temperature
U_b	base velocity $[ms^{-1}]$
U_P	wall velocity $[ms^{-1}]$

Greek symbols

α	Thermal diffusivity, $[m^2s^{-1}]$
β_C	Solutal expansion coefficient
β_T	Thermal expansion coefficient[1/K]
ν	Kinematic viscosity, $[m^2s^{-1}]$
ρ	Fluid density $[kgm^{-3}]$

Subscripts

0	Reference state
opt	Optimal value
TGC	Thermogravitational column
ad	Non dimensional value

earth based research was explained.

Dutrieux et al. [7] reported two independent techniques used to measure positive Soret coefficients. In a first method, the thermo-diffusion coefficient, D_T, was determined by a 5-point sampling process in a thermogravitational column. The second one consisted of velocity measurements of a transient natural convective state. The velocity measurements were obtained by Laser Doppler Velocimetry (LDV).

In a binary fluid mixture subjected to a temperature gradient, a mass fraction gradient appears due to the thermodiffusion or Soret effect. In addition to the usual expression for the mass flux density $\vec{J}_m$ given by the Fick law, a part due to the temperature gradient was added so that:

$$\vec{J}_m \quad \rho D\nabla C \quad \rho D_T C(1 \quad C)\nabla T$$

where D is the mass diffusion coefficient, D_T the thermodiffusion coefficient, ρ the mixture density, T the temperature and C the mass fraction of the constituent of interest.

Mialdun et al. [8] measured diffusion, thermodiffusion and Soret coefficients of water-isopropanol mixtures using three experimental techniques: thermogravitational column, optical beam deflection, and optical digital interferometry. In an analytical and numerical study of the influence of acoustic streaming on species separation in a rectangular cavity, filled with a binary fluid mixture, under microgravity, Charrier-Mojtabi et al. [9] showed that the unicellular flow induced by Eckart streaming may lead to significant species separation. Mahidjiba et al. [10] carried out an analytical and numerical study of the combined action of the Soret effect and shear stress on convection in a horizontal fluid layer filled with a binary mixture. Knobloch and Moore [11] studied the linear stability of Soret-Rayleigh-Bénard convection in a horizontal rectangular cavity. The authors obtained the stability diagram in which the critical Rayleigh number is given as a function of the separation ratio, for a given Lewis number.

In their study of mixed convection in binary mixtures, Khouzam et al. [12] showed that the mass concentration gradient was a function of only two new dimensionless parameters: the mass Rayleigh number Ra_m, and the mass Péclet number, Pe_m in the case where only the upper horizontal wall was moving. Rayleigh-Bénard convection in binary mixtures assuming a thermal diffusion ratio that depends on the local temperature and changes its sign within the cell was studied by Mojtabi et al. [13] and Huke et al. [14]. Yacine et al. [15] studied the Soret-driven convection and separation of binary mixtures in a horizontal porous cavity subjected to cross heat fluxes. They observed that an increase of the heat flux density ratio led to a decrease in the relaxation time.

In the present work, the species separation of a binary fluid mixture under weightlessness is investigated. The two walls $y \quad 0$ and $y \quad H$ of the rectangular cavity move at velocities at velocities $fU_p\,\vec{e_x}$ *and* $U_p\,\vec{e_x}$ respectively and are maintained at uniform and constant temperatures T_h and T_c respectively. The walls $x \quad 0$ and $x \quad L$ are adiabatic and all the walls are impermeable.

An analytical solution is obtained in the case of a shallow cavity. The velocity field and the mass fraction gradient along the x axis are then calculated. Thus, the dimensional mass fraction gradient can be optimized. The analytical results are corroborated by direct 2D numerical simulations. The results obtained, in microgravity, show that the species separation is much greater than that obtained in thermo-gravitational columns.

2. Mathematical formulation

We consider a rectangular cavity of large aspect ratio $B \quad L/H$, where H is the height of the cavity along the y axis and L is the length along the x axis. The cavity is filled with a binary fluid mixture of density and dynamic viscosity μ. The two walls $x \quad 0$ and $x \quad L$ are adiabatic and impermeable. The two other walls $y \quad 0$ and $y \quad H$ are kept at constant and uniform temperature T_h, T_c and move at two opposite but constant imposed velocities $(fU_P\,\vec{e_x},\ U_P\,\vec{e_x})$, $\rho$where f is a dimensionless constant taking the values $f \in [\quad 1,\ 1]$. This constant $f \in [\quad 1,\ 1]$ represents the ratio between the two velocities applied at $y \quad 0$ and $y \quad H$ (see Fig. 1).

The Boussinesq approximation is assumed to be valid; thus, the thermo-physical properties of the binary fluid are constant, except for the density in the buoyancy term, which varies linearly with the local

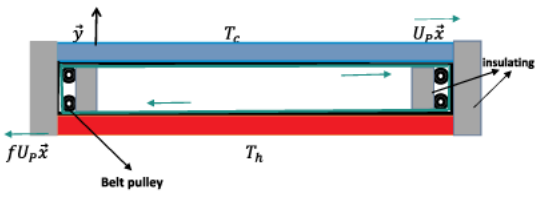

Fig. 1. Schematic diagram of the rectangular cavity.

temperature T and the local mass fraction C of the denser component:

$$\rho \quad \rho_0(1 \quad \beta_T(T \quad T_{ref}) \quad \beta_C(C \quad C_{ref})) \tag{1}$$

where β_T and β_C are respectively the thermal and mass expansion coefficients of the binary fluid defined as follows: $\beta_T \quad \frac{1}{\rho_0}\left(\frac{\partial \rho}{\partial T}\right)_C > 0$, $\beta_C \quad \frac{1}{\rho_0}\left(\frac{\partial \rho}{\partial C}\right)_T < 0$.

The dimensional mathematical formulation of the problem is given by:

$$\begin{cases} \nabla.\vec{V} \quad 0 \\ \dfrac{\partial \vec{V}}{\partial t} + \vec{V}.\nabla\vec{V} \quad \dfrac{\nabla P}{\rho_0} + \nu\nabla^2\vec{V} \\ \dfrac{\partial T}{\partial t} + \vec{V}.\nabla T \quad \alpha\nabla^2 T \\ \dfrac{\partial C}{\partial t} + \vec{V}.\nabla C \quad D\nabla^2 C + D_T\nabla.(C(1-C)\nabla T) \end{cases} \tag{2}$$

Where $\vec{V}$ is the velocity field, inside the rectangular cavity, induced by the walls $y \quad 0$ and $y \quad H$ moving at constant velocity $fU_P\ \vec{e_x}$ and $U_P\ \vec{e_x}$ respectively. Where α is the thermal diffusivity of the binary fluid, ν is the cinematic viscosity, and D_T and D are, respectively, the thermo-diffusion and mass-diffusion coefficient of the denser component.

The corresponding dimensional boundary conditions are:

$$\begin{cases} x \quad 0, x \quad L \ : \vec{V} \quad 0, \ \dfrac{\partial T}{\partial x} \quad \dfrac{\partial C}{\partial x} \quad 0 \\ y \quad 0, H: \ \vec{V} \quad fU_P\vec{e_x}, U_P\vec{e_x}; \ T \quad T_h, T_c, \ D\dfrac{\partial C}{\partial y} + D_T C(1 \ - \ C)\dfrac{\partial T}{\partial y} \quad 0 \end{cases} \tag{3}$$

In many studies, for low species separation, the authors consider that $C(1-C) \approx C_0(1-C_0)$ where, C_0 is the initial mass fraction.

3. Analytical solution in the case of a shallow cavity

In the case of a shallow cavity ($L \gg H$) the parallel flow approximation, used by many previous authors [10], is considered. The solution corresponding to the unicellular flow is as follows:

$$\vec{V_b} \quad U_b(y)\,\vec{e_x}, \quad T_b \quad f(y), \quad C_b \quad mx + h(y) \tag{4}$$

Where $\vec{V_b}$ is the velocity field induced by the sliding the walls $y \quad 0$ and $y \quad H$ at constant velocity $fU_P\ \vec{e_x}$ and $U_P\ \vec{e_x}$ respectively. $\vec{V_b}$ has only the component $U_b(y)\vec{e_x}$ inside the cavity except in the vicinity of the walls $x \quad 0$ and $x \quad L$.

The functions f and h depend of the y component only and m is an unknown constant representing the mass fraction gradient in the x direction.

With these assumptions and for the steady state, the system of equation (2) with the boundary conditions (3) is reduced to the following set of equations, solved using Maple software:

$$\begin{cases} \partial^3 U_b/\partial y^3 \quad 0 \\ \nabla^2 T_b \quad 0 \\ m \ U_b \quad D\nabla^2 C_b \quad D_T C_0(1-C_0)\,\nabla^2 T_b \quad 0 \end{cases} \tag{5}$$

Using the boundary conditions (3), we deduce the expression for the temperature, the velocity and the mass fraction field given by the following expressions:

$$\begin{cases} T_b \quad \dfrac{T_h \quad T_c}{H}y + T_C \\ U_b \quad \dfrac{U_P(f(H \quad y)(H \quad 3y) \quad y(2H \quad 3y)}{H^2} \\ C_b \quad mx + mU_P\left[\dfrac{(f+1)y^4}{4DH^2} \quad \dfrac{(2f+1)y^3}{3DH} + \dfrac{fy^2}{2D} \quad \dfrac{(3f \quad 2)H^2}{60D}\right] + \\ \quad \dfrac{D_T(T_h \quad T_c)C_0(1 \quad C_0)}{DH}\left(y \quad \dfrac{H}{2}\right) \quad \dfrac{mL}{2} + C_0 \end{cases} \tag{6}$$

The value of the mass fraction gradient m:

$$m \quad \frac{35D_T(T_h \quad T_c)C_0(1 \quad C_0)(1 \quad f)U_P}{2H^2U_P^2(2f^2 \quad 3f+2) + 420D^2} \tag{7}$$

is obtained by specifying that the total mass flux of the component of mass fraction C, through any cross section of the rectangular cavity perpendicular to the x axis, is equal to zero:

$$\int_0^H \left(U_bC_b \quad \left(D\frac{\partial C_b}{\partial y} + D_TC_0(1 \quad C_0)\frac{\partial T_b}{\partial y}\right)\right)dy \quad 0 \tag{8}$$

For fixed values of H, the velocity profile $U_b(y)$ depends on the velocity U_P of the planes $y \quad 0$ and $y \quad H$ and on the parameter f.

For $f > 0$, the flow inside the cavity consists of two superimposed contra-rotating cells. This configuration leads to a small mass gradient between the two walls $x \quad 0$ and $x \quad L$, Fig. 2.

4. Optimization of the species separation, S

The species separation of a binary mixture, $S \quad mL$, is defined as the difference in mass fraction of the denser component between the two ends of the cell, $x \quad 0$ and x $\quad L$, with m given by Equation (7). For a given binary mixture, the mass diffusion coefficient D, the thermo-diffusion coefficient D_{TT} and the initial mass fraction of the denser component C_0 are known. The analysis of Equation (7) shows that the species separation decreases when the thickness, H, of the cavity increases. Moreover, the species separation, S, increases when the temperature difference $(T_h \quad T_c)$ increases. Then, two independent control parameters remain, namely the velocity $\boldsymbol{U_P}$ and the parameter f characterizing the ratio of the velocities fU_P and U_P respectively imposed on the walls $y \quad 0$ and $y \quad H$. The optimum separation, S, is obtained by solving the system of equations:

$$\begin{cases} \partial m/\partial U_P \quad 0 \\ \partial m/\partial f \quad 0 \end{cases} \tag{9}$$

Resolution of the algebraic system (9) leads to the values of U_P and f denoted:

$$U_{Popt} \quad \pm\sqrt{30}\,D/H, \quad f_{opt} \quad 1 \tag{10}$$

The optimal velocity U_{Popt}, with respect to the two control parameters U_P and f, depends only of the mass diffusion coefficient and the thickness, H, of the cavity. So, if the mass diffusivity of the binary mixture is known, the experiments can be conducted with this optimal velocity.

Replacing U_P and f by U_{Popt} and f_{opt} respectively in equation (7) yields the expression of maximum mass gradient:

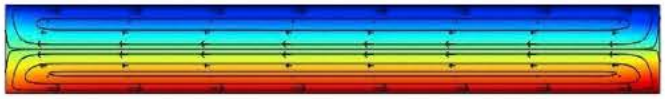

Fig. 2. Stream function and velocity field for water-ethanol binary fluid, $H =$ 2 mm L = 20 mm, and $f = 1$.

$$m_{opt} \quad \sqrt{30}C_0(1 \quad C_0)(T_h \quad T_c)D_T \Big/ 12HD \tag{11}$$

Equation (11) can also be written in the following form:

$$m_{opt} \quad \sqrt{30}C_0(1 \quad C_0)(T_h \quad T_c)S_T \Big/ 12H \tag{12}$$

where $S_T \quad D_T/D$ denotes the Soret coefficient of the binary mixture.

The optimum value of the mass fraction gradient depends only on the thermal gradient in the y direction $(T_h \quad T_c)/H$, once the binary solution characterized by: $C_0(1 \quad C_0)D_T/D$ is known. Equations (10) and (12) show that it is possible to access the indirect measurement of the Soret coefficient S_T by measuring the optimal mass fraction gradient m_{opt}:

$$S_T \quad 12H\, m_{opt} \Big/ \sqrt{30}C_0(1 \quad C_0)(T_h \quad T_c) \tag{13}$$

To make this measurement, it is necessary that the walls be set in motion with the optimal velocity U_{Popt}. The determination of the optimal velocity requiresthe mass diffusivity of the binary mixture to be known.

In what follows, we show that it is also possible to access the measurement of the mass diffusion coefficient D using Equation (7) in the case of $f \quad -1$.

To do this, it is sufficient to carry out two experiments, one with a velocity U_{p1} and the other with velocity U_{p2}, which are not optimal, for the same cell of thickness H, subjected to the same temperature difference $\Delta T \quad (T_h \quad T_c)$.

We then have:

$$m_i \quad 5D_T(T_h \quad T_c)C_0(1 \quad C_0)U_{pi} \Big/ \left(H^2U_{pi}^2 + 30D^2\right) \tag{14}$$

With $i \quad 1$ or 2. The ratio $m_1 \,/\, m_2$ then leads to:

$$m_1/m_2 \quad U_{p1}\left(H^2U_{p2}^2 + 30D^2\right) \Big/ U_{p2}\left(H^2U_{p1}^2 + 30D^2\right) \tag{15}$$

This equation (15) shows that it is possible to determine the mass diffusion coefficient from the measurement of mass fraction gradients m_1 and m_2 obtained for two fixed wall velocities U_{P1} and U_{P2}. From this equation we deduce:

$$D^2 \quad H^2U_{p1}U_{p2}\left(m_2U_{p2} \quad m_1U_{p1}\right)/30\left(m_1U_{p2} \quad m_2U_{p1}\right) \tag{16}$$

For a new mixture with a mass diffusion coefficient easily measured in an earth laboratory, equation (10) indicates the velocity that should be imposed at the two isothermal walls at T_h and T_c. Then, equation (13) allows the calculation of S_T once the mass fraction gradient m has been measured by, for example, taking samples for different values of x.

5. Application to the separation of a water-ethanol binary mixture

5.1. Separation under weightlessness conditions

To illustrate the analytical results obtained in this study, we restrict ourselves to the experimental values of the thermophysics parameters of a water (60.88 wt%)-ethanol (39.12 wt%) solution studied by Platten et al. [16]. The values of the thermophysical properties of this binary solution, at the average temperature $T \quad 22.5°C$ are given in Table 1.

By replacing D, D_T and C_0 by the values presented in Table 1 and for a cavity of thickness $H \quad 2\ 10^{-3}m$, the optimum value of the velocity and the mass fraction gradient following the x axis are obtained as a function of the thermal gradient following the y axis only:

$$\begin{cases} U_{Popt} \quad 4.32\ 10^{-7}\sqrt{30}\Big/2 \approx 1.18\ 10^{-6}\, m\Big/s \\ m_{opt} \quad 0.172(T_h \quad T_c)\ m^{-1} \end{cases} \tag{17}$$

If we take, for example, a temperature difference of 50 °C, the mass fraction gradient for the water-ethanol binary mixture is equal to: 8.60 m^{-1}.

Various direct numerical simulations carried out using the characteristics of the water-ethanol mixture and the same temperature difference in cavities with a thickness of 2 mm and a length between 10 and 90 mm found the same value: $m \quad 8.60\, m^{-1}$ as the one obtained analytically.

For a cavity length $L \quad 20$ mm, the difference between the water mass fraction at the two extremal points $(x \quad 0, y \quad 1\, mm)$ and $(x \quad 20\, mm, y \quad 1\, mm)$ is equal to 0.172. The results of the numerical simulations, from the system of equation (2) and associated boundary conditions (3), led to the same values calculated analytically and numerically when $C(1-C) \quad C_0(1 \ - \ C_0)$, m $\quad 8.60\, m^{-1}$. This numerical result validates the hypothesis $C(1-C) \approx \ C_0(1 \ - \ C_0)$ when the separation of the species is slight.

For $L \quad 90$ mm and $H \quad 2$ mm, when the steady state is reached, the average mass fractions inside the volume near the planes $x \quad 0$ and x L are, respectively, equal to: $\int_y^{\ H}{}_0(\int_x^{\ H}{}_0 C(x,y)dx)dy \quad 0.23$ and $\int_y^{\ H}{}_0$ $(\int_x^{\ L}{}_{L-H} C(x,y)dx)dy \quad 0.996$, which indicates that we have quasi-pure water in the vicinity of $x \quad 90$ mm for $y \in [0, \ 2\, mm]$. In this case, the average mass fractions inside the volume near the plane $x \quad 0$ and $x \quad L$, obtained numerically from the system of equations (2) and (3), when $C_0(1 \ - \ C_0)$ is replaced by $C(1-C)$ are equal to 0.25 and 0.89, respectively. On the other hand, the variation of the mass fraction with x for fixed y is no longer linear. C in the form: $C \quad mx + h\ (y)$ is no longer a solution of the problem studied. Even if the species separation remains almost the same, it is no longer permissible to replace $C(1-C)$ by $C_0(1–C_0)$ when the separation is significant.

For a cavity of greater thickness, $H \quad 5\, mm$, the values of the optimal mass fraction gradient and the associated optimum velocity are equal to:

$$\begin{cases} U_{Popt} \approx 4.73\ 10^{-7}\, m/s \\ m_{opt} \approx 3.45\ m^{-1} \end{cases} \tag{19}$$

Under these conditions, for $L \quad 230\, mm$, and $H \quad 5$ mm, the average mass fraction inside the volume near the planes $x \quad 0$ and $x \quad L$, respectively, are equal to:

$\int_y^{\ H}{}_0(\int_x^{\ H}{}_0 C(x,y)dx)dy \quad 0.22$ and $\int_y^{\ H}{}_0(\int_x^{\ L}{}_{L-H} C(x,y)dx)dy \quad 0.997$ which indicates that we have quasi-pure water in the vicinity of $x \quad 230$ mm and for $y \in [0, \ 5\, mm]$.

To improve the separation process and aware of the non-validity of the hypothesis $C(1-C) \approx \ C_0(1-C_0)$, we carried out simulations by keeping $C(1-C)$ in system (2), but D and D_T remained constant. We found that the separation remains important without m being constant. In the case where D_T changes sign as a function of C, for known D and for a fixed temperature T, the value D_T is calculated from equation (7) for each value of C_0 that will be varied for determine the value of C_0 leading to $D_T \quad 0$. Onset of free convection in solutions with variable Soret coefficients was studied by Mojtabi et al. [13].

5.2. Comparison with the separation inside a thermogravitational column under gravity conditions

Using the parallel flow hypothesis presented in paragraph 4 and

Table 1
Properties for a water (60.88 wt%) - ethanol (39.12 wt%) mixture at a mean temperature of 22.5°C.

$D[m^2s^{-1}]$	$D_T[m^2s^{-1}K^{-1}]$	β_C	$\beta_T[K^{-1}]$	$\alpha[m^2s^{-1}]$	$\rho_0[kg.m^{-3}]$	$\nu[m^2s^{-1}]$
4.3210^{-10}	$1.37\ 10^{-12}$	− 0.212	7.8610^{-4}	10^{-7}	935.17	2.71610^{-6}

assuming the forgotten effect proposed by Furry et al. [17], we obtain the velocity field and the vertical mass fraction gradient in a thermogravitational column (TGC) of thickness H, height L and subjected to a temperature difference ΔT $(T_h$ $T_c)$:

$$\begin{cases} W(x) & \beta_T g(T_h \quad T_c)x(H \quad x)(H \quad 2x)/12H\nu \\ m_{TGC} & 504H^2\beta_T D_T \nu g(T_h \quad T_c)^2 C_0(1 \quad C_0)/\left(H^6\beta_T^2 g^2(T_h \quad T_c)^2 + 362880D^2\nu^2\right) \end{cases} \tag{20}$$

where $\vec{W}$ $W(x)\vec{e_y}$.

By replacing β_T, ν, D, D_T and C_0 by their values indicated in Table 1 and for a TGC of thickness H $2.10^{-3}m$ and $(T_h$ $T_c)$ $50\,^{\circ}C$, where g is the gravitational intensity, the maximum value of the vertical velocity and the associated mass fraction gradient along the y axis is obtained:

$$\begin{cases} W_{max} \approx 4.55\ 10^{-3}\ m/s \\ m_{TGC} \approx 3.62\ 10^{-3} m^{-1} \end{cases} \tag{21}$$

Thus, the mass fraction gradient, $m_{opt} \approx 8.62\ m^{-1}$ obtained in microgravity is 2381 times greater than the $m_{TGC} \approx 3.6210^{-3} m^{-1}$ obtained in a thermogravitational column of the same thickness and subjected to the same temperature gradient. For the thermogravitational column, at each temperature difference, there is a corresponding optimum of separation for a particular value of the thickness H_{opt}. This thickness H_{opt} is around $10^{-4} m^{-1}$. For a thickness $H < H_{opt,}$ m_{TGC} decreases rapidly. In order to obtain a species separation of the binary mixture that is of the same order of magnitude as that obtained in microgravity, for $(T_h$ $T_c)$ $50^{\circ}C$ and H $2\ 10^{-3}\ m$, a thermogravitational column of thickness H $3.10^{-4} m$ should be considered. In this case, the maximum values of velocity and the vertical mass fraction gradient, in the thermogravitational column are:

$$\begin{cases} W_{max} \approx 1.11\ 10^{-3}\ m/s \\ m_{TGC} \approx 7.12\ m^{-1} \end{cases} \tag{22}$$

For this small column thickness, the amount of separated component is very small and, furthermore, measuring the mass fraction of one of the components at different points along the height of the column is a very difficult task.

5.3. Results in dimensionless form

As indicated in Equation (7) the dimensional mass fraction gradient m depends on H, Up, $(T_h$ $T_c)$, $C_0(1$ $C_0)$, D_T, D and f. If we denote respectively by H, H^2/α, α/H, $(T_h$ $T_c)$ and ΔC $(T_h$ $T_c)C_0(1$ $C_0)D_T/D$ the reference scales of length, time, velocity, temperature and mass fraction, respectively, we deduce the non-dimensional expression of the mass fraction gradient and the associated velocity.

5.3.1. Results under weightlessness conditions

The mass fraction gradient is given by:

$$m^{ad} \quad \frac{35PeLe(1 \quad f)}{2Pe^2Le^2\left(2f^2 \quad 3f+2\right)+420} \quad \frac{35Pe_m(1 \quad f)}{2Pe_m^2\left(2f^2 \quad 3f+2\right)+420} \tag{23}$$

where Pe U_P H/α, Le α/D and Pe_m denote respectively the Péclet, the Lewis and the mass Péclet numbersPe_m $Pe\ Le$ U_P H/D. For the water-ethanol binary mixture defined in Table 1, the non-dimensional number involved in the expression of m has the value:

Le 231.5 . The non-dimensional mass fraction gradient, m^{ad}, depends on only the two parameters Pe_m and f (eq.24). The optimal value of m_{opt}^{ad} is obtained for Pe_m $\sqrt{30}$ and f 1 and leads to:

$$m_{opt}^{ad} \quad \sqrt{30}/12 \approx 0.456 \tag{24}$$

By again taking a cavity of thickness H 2 mm filled with the same binary water-ethanol solution, it is possible to deduce the value of dimensional velocity from $Pe_{m,\ opt}$:

$$Pe_{m,\ opt} \quad U_P\ H/D \quad 0.463.10^7 U_P \quad \sqrt{30} \quad > \quad U_P \quad 1.18\ 10^{-6} m/s \tag{25}$$

This agrees with the value given by Equation (10).

5.3.2. Comparison with the separation obtained with a thermogravitational column

The velocity and the mass fraction gradient are given by:

$$\begin{cases} W(x) & Ra(1 \quad x)(1 \quad 2x)x/12 \\ m_{TGC} & 504RaLe/\left(Le^2Ra^2 + 362880\right) \end{cases} \tag{26}$$

where Ra $g\beta_T(T_h \quad T_c)H^3/\alpha\nu$ and Le α/D denote the thermal Rayleigh number and the Lewis number, respectively. For the water-ethanol binary mixture defined in Table 1, the non-dimensional number involved in the expression of m_{TGC} is: Le 231.5. The non-dimensional mass fraction gradient m_{TGC}, depends only on the two parameters $Ra$and Le.

The non-dimensional separation is defined as S $m_{TGC}L/H$.

Thus, the dimensional value of the separation between the two horizontal surfaces y 0 and y L of the vertical column is given by:

$$S^r \quad m_{TGC}^r L \quad C_0(1 \quad C_0)D_T(T_h \quad T_c)m_{TGC}L/DH \tag{27}$$

where m_{TGC}^r is the dimensional mass fraction gradient for the TGC. Using the physical values of the water-ethanol binary mixture given in Table 1, we obtain:

$$m_{TGC}^r \quad 3.621\ 10^{-3} m^{-1} \tag{28}$$

The optimal value of m_{TGC} is obtained for the optimal value of thermal Rayleigh number Ra_{opt} $\frac{72\sqrt{70}}{\sqrt{Le}} \approx 39.59$ and leads to: m_{TGC}^{opt} $\sqrt{70}/20\sqrt{Le} \approx 0.275$. This optimum is associated with a very small thickness of the cavity, H $1.09\ 10^{-4}\ m$. The mass of the separated constituent is then very weak.

6. . 2D numerical simulations

The dimensionless set of equations and boundary conditions obtained from systems (2) and (3) with the reference scales defined in § 5.3 was solved numerically using a finite element code (Comsol Multiphysics), with rectangular mesh. The time-dependent solver and the set of equations (incompressible Navier-Stokes, thermal and mass diffusion equations) in transient form were used. The condition of conservation of the average mass fraction in the cavity was imposed on each of the iterations. Direct numerical simulations were performed for cavities of the same height H 2 mm with lengths L 20, 40, 70 and 90 mm, which correspond to aspect ratios of 10, 20 , 35 and 45, respectively . The quadrangle spatial resolutions were 20 200 up to 30 700 for the cell with aspect ratio 45 (see Fig. 3).

For f 1 and for the water-ethanol mixture defined in Table 1, Le 231.5, the analytical calculations lead to m_{opt}^{ad} 0.456, equation

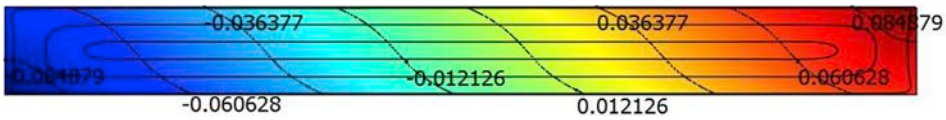

Fig. 3. Stream function, iso-mass fraction lines and mass fraction field (in colour) for $f =$ 1, $\Delta T = 20$, $L = 70\,mm$, $H = 2\,mm$.

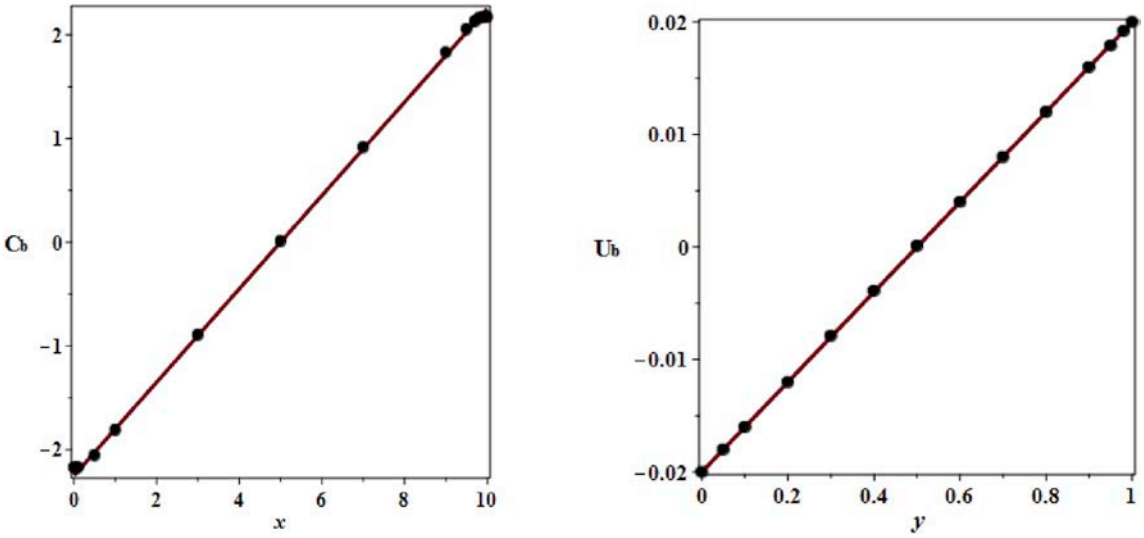

Fig. 4. (a) (on the left) Mass fraction $C_b\left(x,\ y=\frac{1}{2}\right)$ and *(b)* (on the right) velocity $U_b(x=5,y)$ for $Pe = 0.02$ and $A = 107$.

(25). The plot of the mass fraction C_b as a function of x for y 1/ 2 (Fig. 4 (a)) and the plot of the velocity $U_b(y)$ in x 5 (Fig. 4 (b)) for Pe 0.02 and A 10, show very good agreement between the analytical calculations (red line) and the direct numerical simulation results (dark points).

7. Conclusion

In this work, we show the possibility of greatly improving the species separation of a binary mixture in weightlessness by using a rectangular cavity that has two opposite walls of great length, moving with equal optimal velocities in opposite directions, U_{Popt} *and* U_{Popt}. There is a temperature difference of ΔT between these walls.

In this new configuration, the species separation is several orders of magnitude greater than that obtained in thermo-gravitational columns (TGC).

It also emerges from this study that it is possible to carry out experiments in microgravity in order to directly measure the mass diffusion coefficient of a binary mixture. To do this, it is sufficient to perform two experiments, one with wall velocities U_{p1} *and* U_{p1} and the other with wall velocities U_{p2} and U_{p2} imposed on the mobile walls, and to measure the values of the mass fraction gradients m_1 and m_2 associated with each of these two experiments. Determining the ratio $m_1/\ m_2$ allows the mass diffusion coefficient D, to be calculated from Equation (17).

The measurement of the mass diffusion coefficient of a binary mixture is generally carried out in ground based experiments. Once D has been measured on the ground, Equation (10) allows the optimum velocity, U_{Popt} to be calculated for a cell of thickness H. When the optimal mass fraction m_{opt} has been measured for a given temperature difference ΔT, D_T or S_T can be calculated from equation (11) or (13) respectively. In order to determine D_T or S_T in microgravity, for a given binary mixture, the following procedure will be used:

- several identical cells will be built,
- a different ΔT will be imposed on each cell,
- for each ΔT, the U_{Popt} will be calculated and the m_{opt} will be measured.

Declaration of competing interest

None.

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